Dressing up for length gauge: Aspects of a debate in quantum optics

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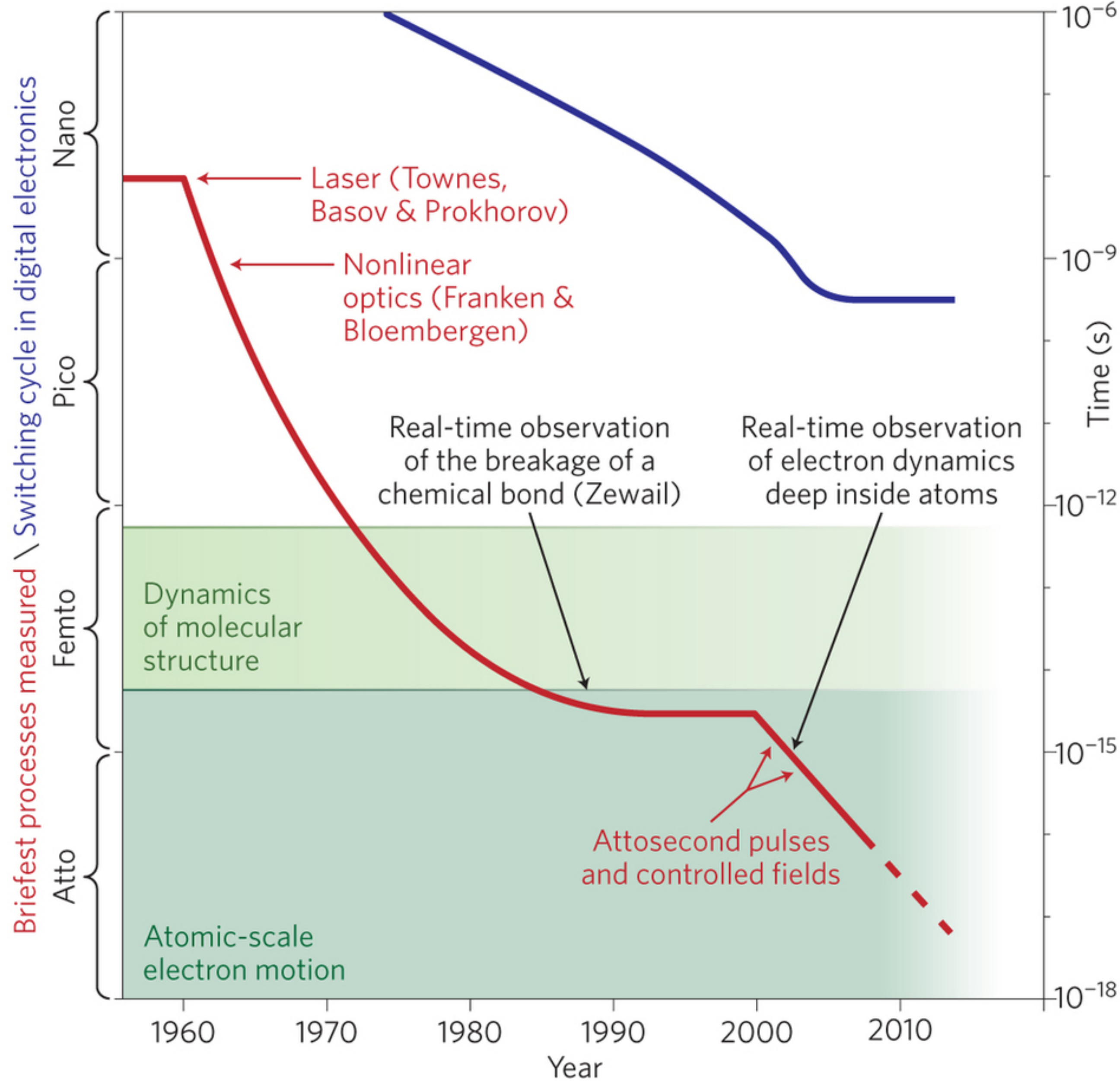
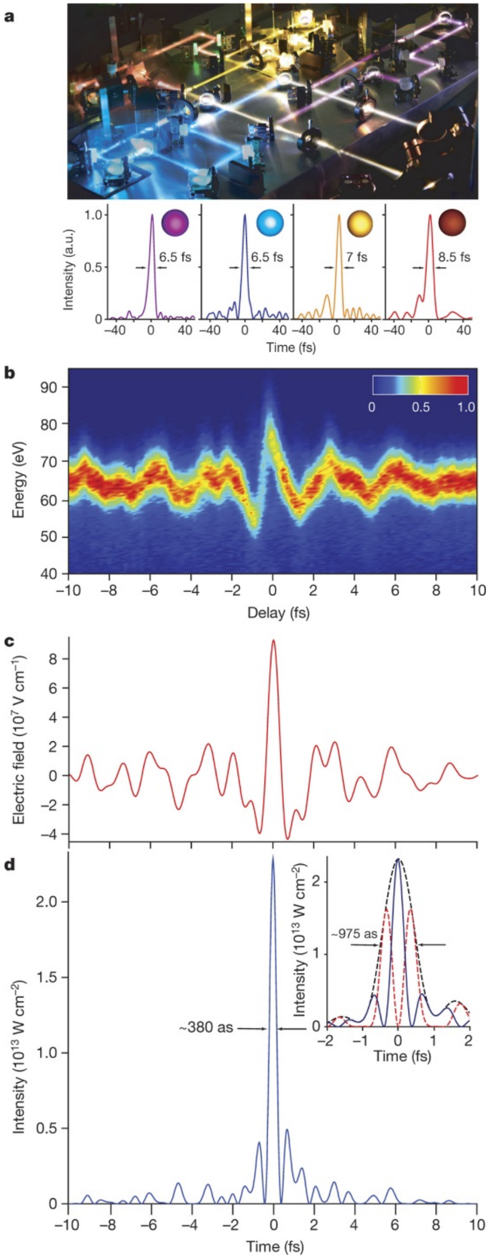
Agenda:

- Attosecond spectroscopy
- Electron detachment in strong fields
- Optical properties in chiral materials
- Quantum optics in Coulomb gauge
- Dipole approximation
- Schrödinger-Maxwell system in velocity gauge and length gauge
- Problems with length gauge versus velocity gauge
- Reasons for failure of analytic equivalence of velocity gauge and length gauge

Evolution of ultrafast science and digital electronics

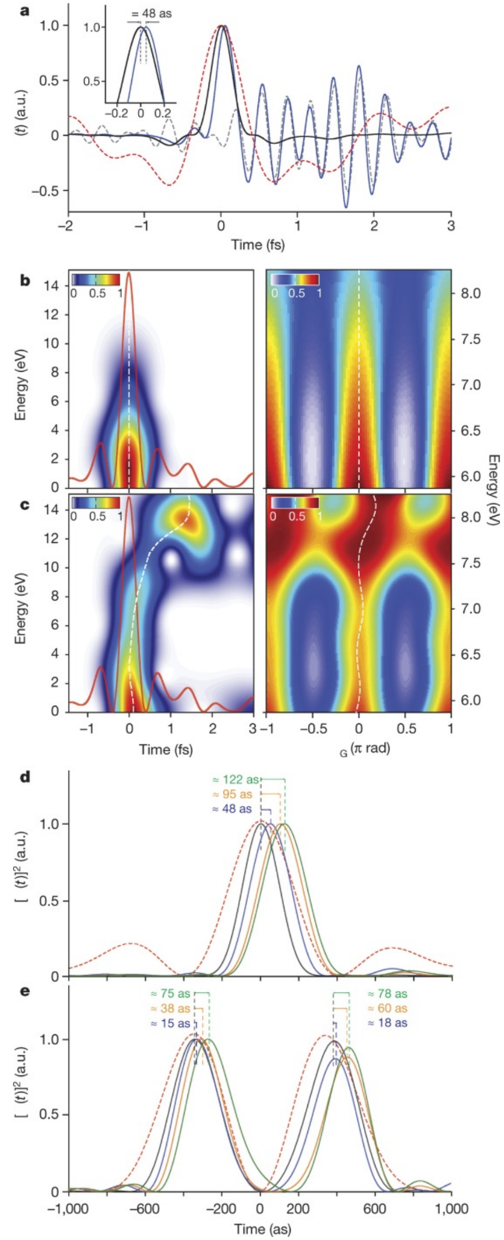


F. Krausz and M. I. Stockman *Nature Photonics* **8,** 205-213 (2014) doi:10.1038/nphoton.2014.28



Synthesis of an optical attosecond pulse

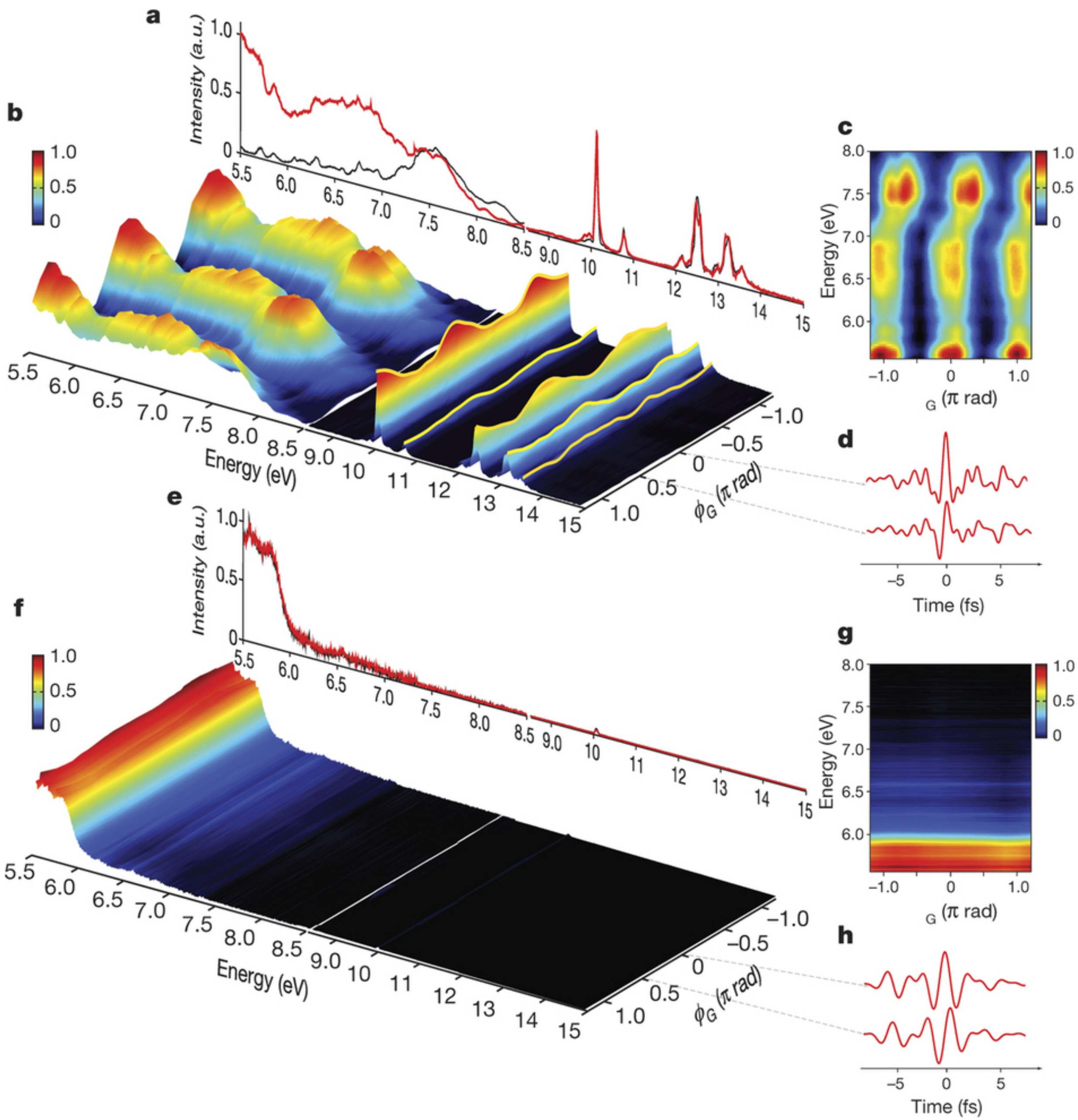
Nonlinear response of bound electrons of Kr to an optical attosecond pulse



M. Th. Hassan *et al. Nature* **530,** 66–70 (2016) doi:10.1038/nature16528

nature

Attosecond control of bound electrons in Kr



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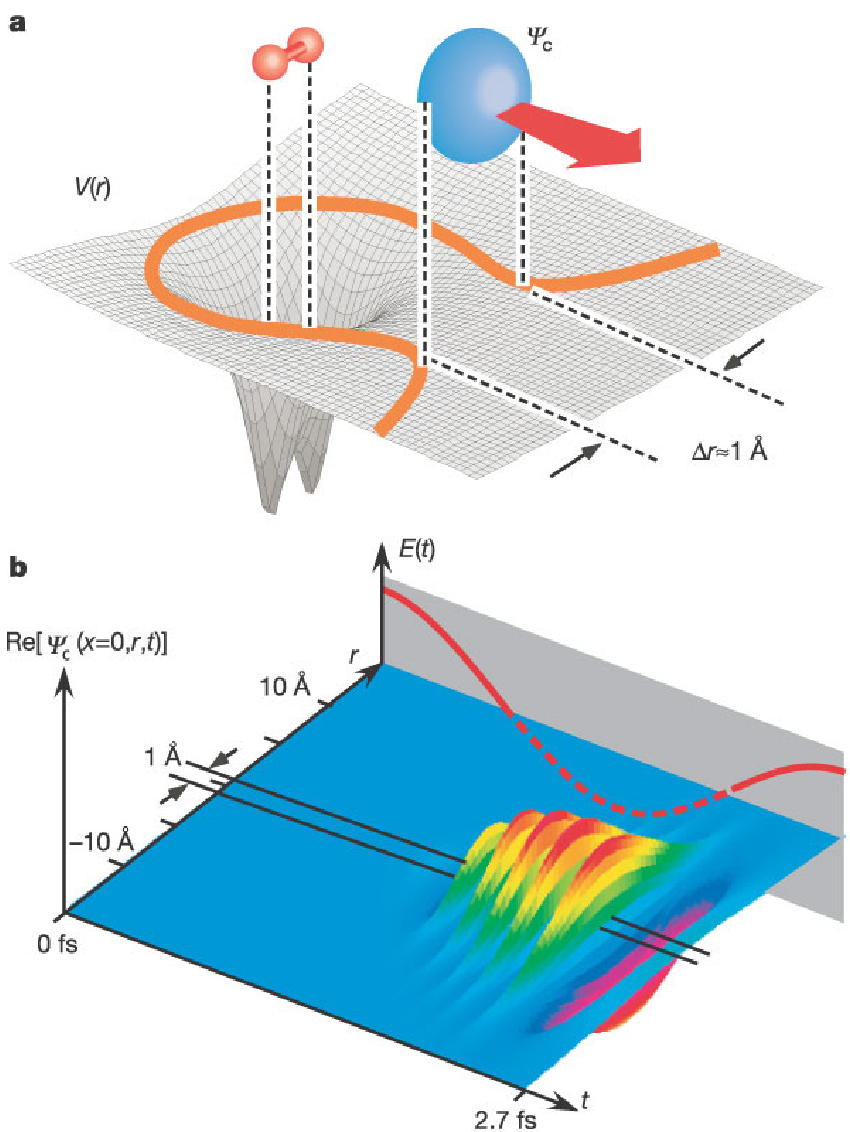
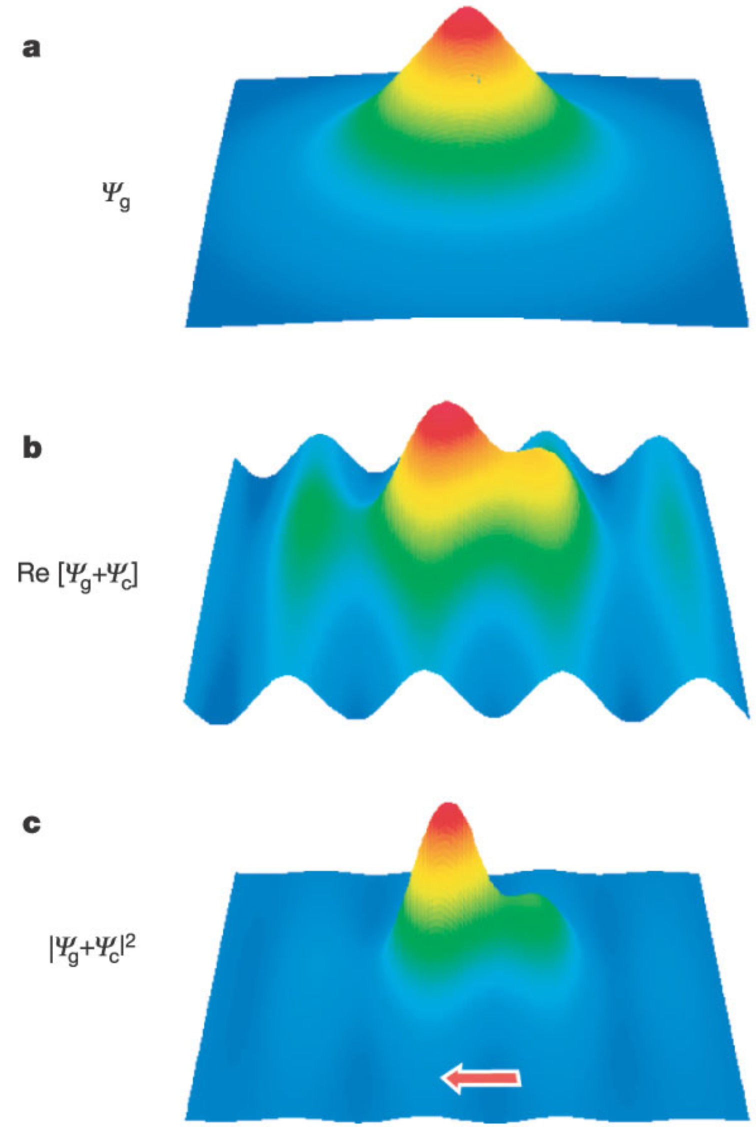
Another application: electron detachment from N_2 and recollision in strong laser field





J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J. C. Kieffer, P. B. Corkum & D. M. Villeneuve, *Nature* **432**, 867-871 (2004) doi:10.1038/nature03183

Reconstruction of highest occupied molecular orbital in N_2 and comparison with theory

J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J. C. Kieffer, P. B. Corkum & D. M. Villeneuve, *Nature* **432**, 867-871 (2004)
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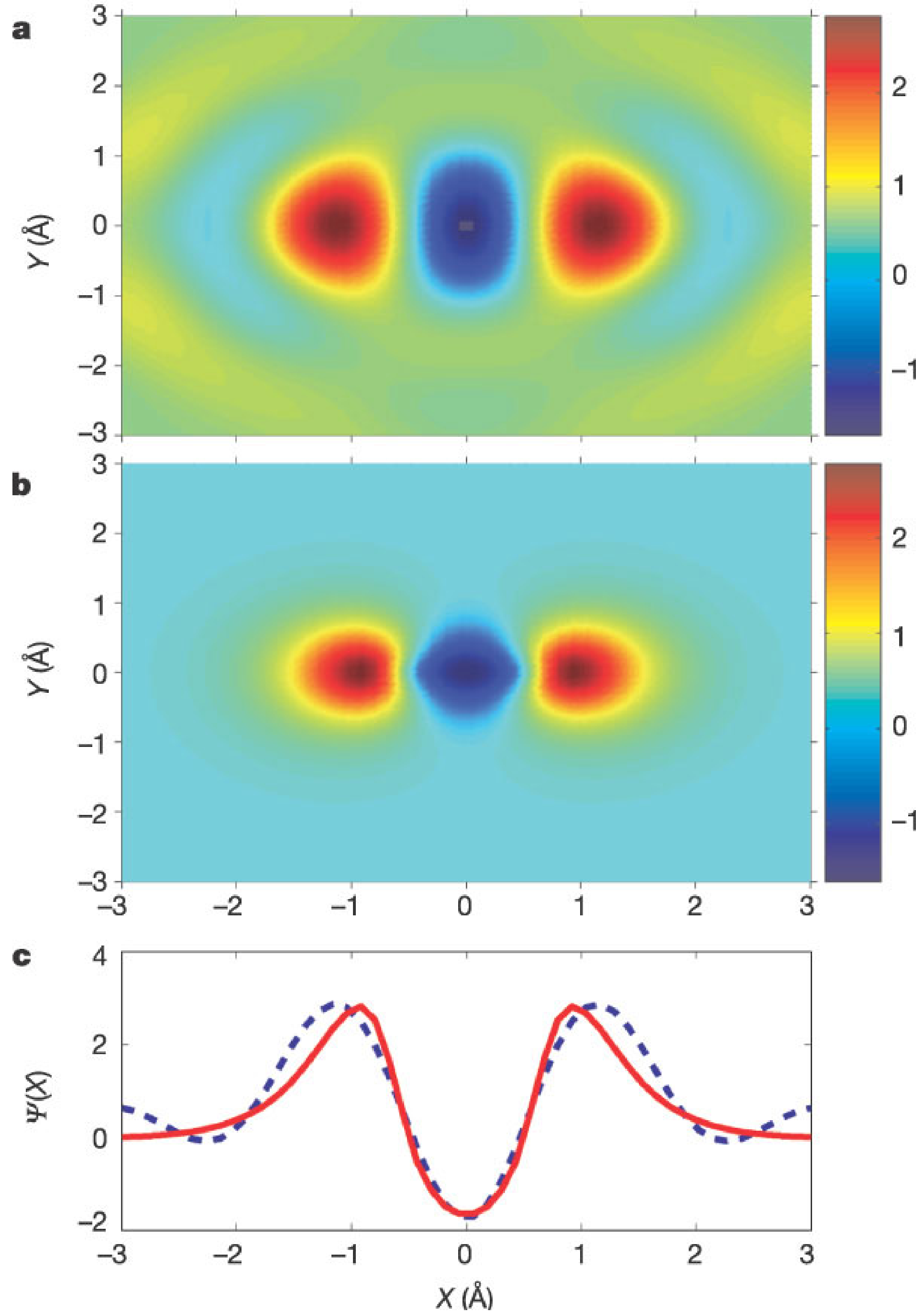

The mathematical analysis of these experiments is ultimately based on the quantum optics Hamiltonian in Coulomb gauge

$$\nabla \cdot \boldsymbol{A}(\boldsymbol{x},t) = 0,\ \boldsymbol{E}_\perp(\boldsymbol{x},t) = -\,\partial \boldsymbol{A}(\boldsymbol{x},t)/\partial t,\ \boldsymbol{B}(\boldsymbol{x},t) = \nabla \times \boldsymbol{A}(\boldsymbol{x},t),$$

$$\begin{aligned}
H =& \int d^3\boldsymbol{x} \sum_a \frac{1}{2m_a}\left(\hbar^2 \nabla\psi_a^+(\boldsymbol{x},t)\cdot\nabla\psi_a(\boldsymbol{x},t) + q_a^2\psi_a^+(\boldsymbol{x},t)\boldsymbol{A}^2(\boldsymbol{x},t)\psi_a(\boldsymbol{x},t)\right)\\
&+ \int d^3\boldsymbol{x} \sum_a \frac{iq_a\hbar}{2m_a}\boldsymbol{A}(\boldsymbol{x},t)\cdot\left(\psi_a^+(\boldsymbol{x},t)\cdot\nabla\psi_a(\boldsymbol{x},t) - \nabla\psi_a^+(\boldsymbol{x},t)\cdot\psi_a(\boldsymbol{x},t)\right)\\
&+ \int d^3\boldsymbol{x}\left(\frac{\epsilon_0}{2}\boldsymbol{E}_\perp^2(\boldsymbol{x},t) + \frac{1}{2\mu_0}\boldsymbol{B}^2(\boldsymbol{x},t) + \sum_a \psi_a^+(\boldsymbol{x},t)V_a(\boldsymbol{x},t)\psi_a(\boldsymbol{x},t)\right)\\
&+ \iint d^3\boldsymbol{x}\,d^3\boldsymbol{x}'\,\frac{1}{2}\sum_{aa'}\psi_a^+(\boldsymbol{x},t)\psi_{a'}^+(\boldsymbol{x}',t)V_{aa'}(\boldsymbol{x}-\boldsymbol{x}',t)\psi_{a'}(\boldsymbol{x}',t)\psi_a(\boldsymbol{x},t),
\end{aligned}$$

$$\frac{q_a q_{a'}}{4\pi\epsilon_0|\boldsymbol{x}-\boldsymbol{x}'|} \subseteq V_{aa'}(\boldsymbol{x}-\boldsymbol{x}',t)$$

Quantum optics with photons in the sub-keV energy is conveniently described in dipole approximation, $\boldsymbol{A}(\boldsymbol{x},t) \approx \boldsymbol{A}(t)$, since photons with wavelengths exceeding 10 nm cannot resolve atomic or molecular length scales.

→ The term for interaction between matter and light takes the "velocity form"

$$H_{Iv} = \int d^3\boldsymbol{x} \sum_a \frac{iq_a\hbar}{2m_a}\boldsymbol{A}(t)\cdot\left(\psi_a^+(\boldsymbol{x},t)\cdot\nabla\psi_a(\boldsymbol{x},t) - \nabla\psi_a^+(\boldsymbol{x},t)\cdot\psi_a(\boldsymbol{x},t)\right)$$

$$+\int d^3\boldsymbol{x}\sum_a \frac{q_a^2}{2m_a}\psi_a^+(\boldsymbol{x},t)\boldsymbol{A}^2(t)\psi_a(\boldsymbol{x},t)$$

The Hamiltonian and equations of motion in velocity form are invariant under residual gauge transformations

$$\psi_a(\boldsymbol{x},t) \to \psi_a'(\boldsymbol{x},t) = \exp\left[iq_a\left(\boldsymbol{a}(t)\cdot\boldsymbol{x} + b(t)\right)/\hbar\right]\psi_a(\boldsymbol{x},t),$$

$$\boldsymbol{A}(t) \to \boldsymbol{A}'(t) = \boldsymbol{A}(t) + \boldsymbol{a}(t),$$

$$V_{a_1a_2}(\boldsymbol{x}_1 - \boldsymbol{x}_2, t) \to V'_{a_1a_2}(\boldsymbol{x}_1 - \boldsymbol{x}_2, t) = V_{a_1a_2}(\boldsymbol{x}_1 - \boldsymbol{x}_2, t),$$

$$V_a(\boldsymbol{x},t) \to V_a'(\boldsymbol{x},t) = V_a(\boldsymbol{x},t) - q_a\dot{\boldsymbol{a}}(t)\cdot\boldsymbol{x} - q_a\dot{b}(t)$$

In particular the gauge transformation

$$\psi_a(\boldsymbol{x},t) \to \psi'_a(\boldsymbol{x},t) = \exp[-iq_a\boldsymbol{x}\cdot\boldsymbol{A}(t)/\hbar]\psi_a(\boldsymbol{x},t),$$

$$\boldsymbol{A}(t) \to \boldsymbol{A}'(t) = 0,\ \ V_{a_1a_2}(\boldsymbol{x}_1-\boldsymbol{x}_2,t) \to V'_{a_1a_2}(\boldsymbol{x}_1-\boldsymbol{x}_2,t) = V_{a_1a_2}(\boldsymbol{x}_1-\boldsymbol{x}_2,t),$$

$$V_a(\boldsymbol{x},t) \to V'_a(\boldsymbol{x},t) = V_a(\boldsymbol{x},t) + q_a\boldsymbol{x}\cdot d\boldsymbol{A}(t)/dt = V_a(\boldsymbol{x},t) - q_a\boldsymbol{x}\cdot\boldsymbol{E}\,(t)$$

transforms the interaction term between matter and light into the “length form”

$$H_{Il} = -\int d^3\boldsymbol{x}\sum_a q_a\psi_a^+(\boldsymbol{x},t)\boldsymbol{x}\cdot\boldsymbol{E}(t)\psi_a(\boldsymbol{x},t)$$

The Hamiltonian and equations of motion in length form are invariant under residual gauge transformations

$$\psi_a(\boldsymbol{x},t) \to \psi'_a(\boldsymbol{x},t) = \exp[iq_ac(t)/\hbar]\psi_a(\boldsymbol{x},t),$$

$$V_a(\boldsymbol{x},t) \to V'_a(\boldsymbol{x},t) = V_a(\boldsymbol{x},t) - q_a\dot{c}(t)$$

The Heisenberg equations of motion in velocity gauge are

$$i\hbar\,\partial\psi_a(\boldsymbol{x},t)/\partial t = -\left[\hbar\boldsymbol{\nabla} - iq_a\boldsymbol{A}(t)\right]^2\psi_a(\boldsymbol{x},t)/2m_a + V_a(\boldsymbol{x},t)\psi_a(\boldsymbol{x},t)$$
$$+\sum_b\int d^3\boldsymbol{x}'\psi_b^+(\boldsymbol{x}',t)\,V_{ab}(\boldsymbol{x}-\boldsymbol{x}',t)\psi_b(\boldsymbol{x}',t)\psi_a(\boldsymbol{x},t)$$

The Heisenberg equations of motion in length gauge are

$$i\hbar\,\partial\psi_a(\boldsymbol{x},t)/\partial t = -\hbar^2\Delta\,\psi_a(\boldsymbol{x},t)/2m_a + V_a(\boldsymbol{x},t)\psi_a(\boldsymbol{x},t) - q_a\boldsymbol{x}\cdot\boldsymbol{E}(t)\psi_a(\boldsymbol{x},t)$$
$$+\sum_b\int d^3\boldsymbol{x}'\psi_b^+(\boldsymbol{x}',t)\,V_{ab}(\boldsymbol{x}-\boldsymbol{x}',t)\psi_b(\boldsymbol{x}',t)\psi_a(\boldsymbol{x},t)$$

These systems are gauge equivalent, and yet they yield very different results!

Example: The differential electron-photon scattering cross section in velocity gauge is (with $ck' = ck - \omega_{n',n} = ck - \omega_{n'} + \omega_n$)

$$\frac{d\sigma}{d\Omega} = \frac{\alpha_S^2 k'}{c^2 k}\left|\frac{\hbar}{m_e}\delta_{n,n'}\,\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\boldsymbol{\epsilon}(\boldsymbol{k}) + M_{fi}\right|^2$$

$$M_{fi} = \sum_{n''}\omega_{n',n''}\omega_{n'',n}\left(\frac{\langle n'|\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\mathbf{x}|n''\rangle\langle n''|\boldsymbol{\epsilon}(\boldsymbol{k})\cdot\mathbf{x}|n\rangle}{\omega_{n'',n} - ck - i\varepsilon} + \frac{\langle n'|\boldsymbol{\epsilon}(\boldsymbol{k})\cdot\mathbf{x}|n''\rangle\langle n''|\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\mathbf{x}|n\rangle}{\omega_{n'',n} + ck' - i\varepsilon}\right)$$

while length gauge yields the original Kramers-Heisenberg formula

$$\frac{d\sigma}{d\Omega} = \alpha_S^2 c^2 k k'^3\left|\widetilde{M}_{fi}\right|^2$$

$$\widetilde{M}_{fi} = \sum_{n''}\left(\frac{\langle n'|\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\mathbf{x}|n''\rangle\langle n''|\boldsymbol{\epsilon}(\boldsymbol{k})\cdot\mathbf{x}|n\rangle}{\omega_{n'',n} - ck - i\varepsilon} + \frac{\langle n'|\boldsymbol{\epsilon}(\boldsymbol{k})\cdot\mathbf{x}|n''\rangle\langle n''|\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\mathbf{x}|n\rangle}{\omega_{n'',n} + ck' - i\varepsilon}\right)$$

Example: The differential electron-photon scattering cross section in velocity gauge is (with $ck' = ck - \omega_{n',n} = ck - \omega_{n'} + \omega_n$)

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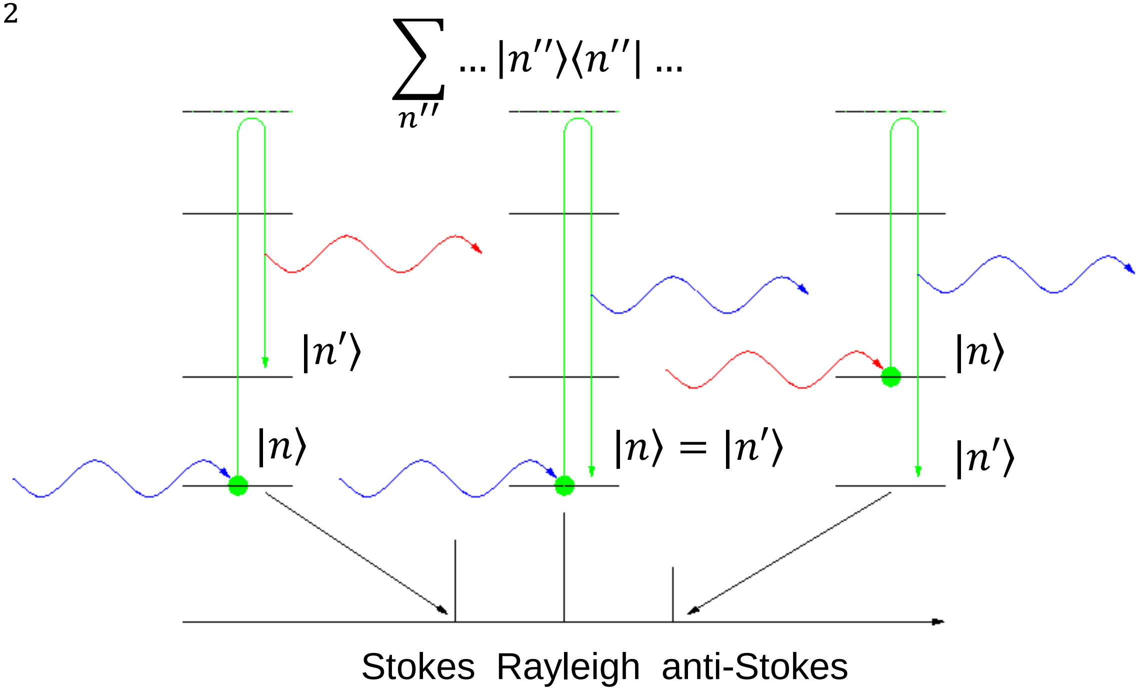
$$\frac{d\sigma}{d\Omega} = \frac{\alpha_S^2 k'}{c^2 k}\left|\frac{\hbar}{m_e}\delta_{n,n'}\,\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\boldsymbol{\epsilon}(\boldsymbol{k}) + M_{fi}\right|^2$$



while length gauge yields the original Kramers-Heisenberg formula

$$\frac{d\sigma}{d\Omega} = \alpha_S^2 c^2 k k'^3 \left|\widetilde{M}_{fi}\right|^2$$

$$\widetilde{M}_{fi} = \sum_{n''}\left(\frac{\langle n'|\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\mathbf{x}|n''\rangle\langle n''|\boldsymbol{\epsilon}(\boldsymbol{k})\cdot\mathbf{x}|n\rangle}{\omega_{n'',n} - ck - i\varepsilon} + \frac{\langle n'|\boldsymbol{\epsilon}(\boldsymbol{k})\cdot\mathbf{x}|n''\rangle\langle n''|\boldsymbol{\epsilon}'(\boldsymbol{k}')\cdot\mathbf{x}|n\rangle}{\omega_{n'',n} + ck' - i\varepsilon}\right)$$

Select instances of strong differences of velocity gauge and length gauge results

Electron detachment from atoms or ions in strong fields	Angular and momentum distributions of emitted electrons; length gauge in general preferred	D. Bauer, Milošević & Becker 2005; Bergues *et al.* 2007; Zhang & Nakajima 2007; J.H. Bauer 2016
Optical properties of graphene	Photo-induced carriers and conductance; velocity gauge preferred	Dong, Han & Xu 2014
Optical properties of chiral molecules	Line strengths and absorption cross sections	Kamiński *et al.* 2015; Friese & Ruud 2016

Sources of differences between velocity gauge and length gauge

1. First order matrix elements are usually equivalent due to the relations

$$\hbar\mathbf{p} = \mathrm{i}m[H,\mathbf{x}] \;\rightarrow\; \langle f|\mathbf{p}|i\rangle = \mathrm{i}m\omega_{fi}\langle f|\mathbf{x}|i\rangle \rightarrow\; \langle f|\mathbf{p}|i\rangle = \pm\mathrm{i}mck\langle f|\mathbf{x}|i\rangle$$

   if $\omega_{fi} \equiv \omega_f - \omega_i = \pm ck$.

   However, for short pulses of duration Δt the energy preserving factor in transition matrix elements is replaced by the Dirichlet kernel

$$\frac{\sin\left(\left(\omega_{fi} \mp ck\right)\Delta t/2\right)}{\pi\left(\omega_{fi} \mp ck\right)}$$

→ This yields discrepancies between velocity gauge and length gauge at the several percent to several ten percent level for sub-femtosecond pulses.

2. *The first order relation does not work for electron detachment from atoms or ions in strong fields*, since the initial and final states in those experiments are eigenstates of different Hamiltonians: Coulomb potentials dominate for bound electron states, but external electric fields from intense laser pulses dominate for detached electron states.

Sources of differences between velocity gauge and length gauge (continued)

3. The gauge transformation

$$|\psi(t)\rangle \rightarrow |\psi'(t)\rangle = \exp[-\mathrm{i}q\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar]|\psi(t)\rangle$$

$$\boldsymbol{A}(t) \rightarrow \boldsymbol{A}'(t) = 0,$$

$$V(\mathbf{x},t) \rightarrow V'(\mathbf{x},t) = V(\mathbf{x},t) + q\mathbf{x}\cdot d\boldsymbol{A}(t)/dt = V(\mathbf{x},t) - q\mathbf{x}\cdot\boldsymbol{E}\,(t)$$

does *not* generate a unitary transformation of the Hamiltonians

$$H_v(t) = \frac{[\mathbf{p} - q\boldsymbol{A}(t)]^2}{2m} + V(\mathbf{x},t)$$

$$\rightarrow\ H_l(t) = \exp[-\mathrm{i}q\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar][H_v(t) + q\mathbf{x}\cdot d\boldsymbol{A}(t)/dt]\exp[\mathrm{i}q\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar]$$
$$= \frac{\mathbf{p}^2}{2m} + V(\mathbf{x},t) + q\mathbf{x}\cdot\frac{d\boldsymbol{A}(t)}{dt}$$

$\rightarrow$ the matrix elements are different

$$\langle\phi'(t)|H_l(t)|\psi'(t)\rangle = \langle\phi(t)|H_v(t)|\psi(t)\rangle + q\langle\phi(t)|\mathbf{x}|\psi(t)\rangle\cdot d\boldsymbol{A}(t)/dt$$

Contrary to an ordinary gauge transformation, the transformation

$$|\psi_a(t)\rangle \to |\psi'_a(t)\rangle = \exp[-\mathrm{i}q_a\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar]|\psi_a(t)\rangle$$

$$V_a(\mathbf{x},t) \to V'_a(\mathbf{x},t) = V_a(\mathbf{x},t) + q_a\mathbf{x}\cdot d\boldsymbol{A}(t)/dt$$

does not necessarily change the gauge fields, and can therefore be separately applied for each particle species:

$$H_{vl}(t) = \sum_{a=1}^{n_v}\left(\frac{[\mathbf{p}_{\boldsymbol{a}} - q_a\boldsymbol{A}(t)]^2}{2m_a} + V_a(\mathbf{x},t)\right) + \sum_{a=1}^{n_l}\left(\frac{\mathbf{p}_a^2}{2m_a} + V_a(\mathbf{x},t) + q_a\mathbf{x}\cdot\frac{d\boldsymbol{A}(t)}{dt}\right)$$

Within a Hartree approximation for many-particle states, the choice between length gauge and velocity gauge can even be separately imposed for each orbital or particle in the system.

Conclusions (or rather: observations)

- The transition from velocity gauge to length gauge does not generate a unitary transformation of the Hamiltonian.

- The transition can be done selectively for different particle species.

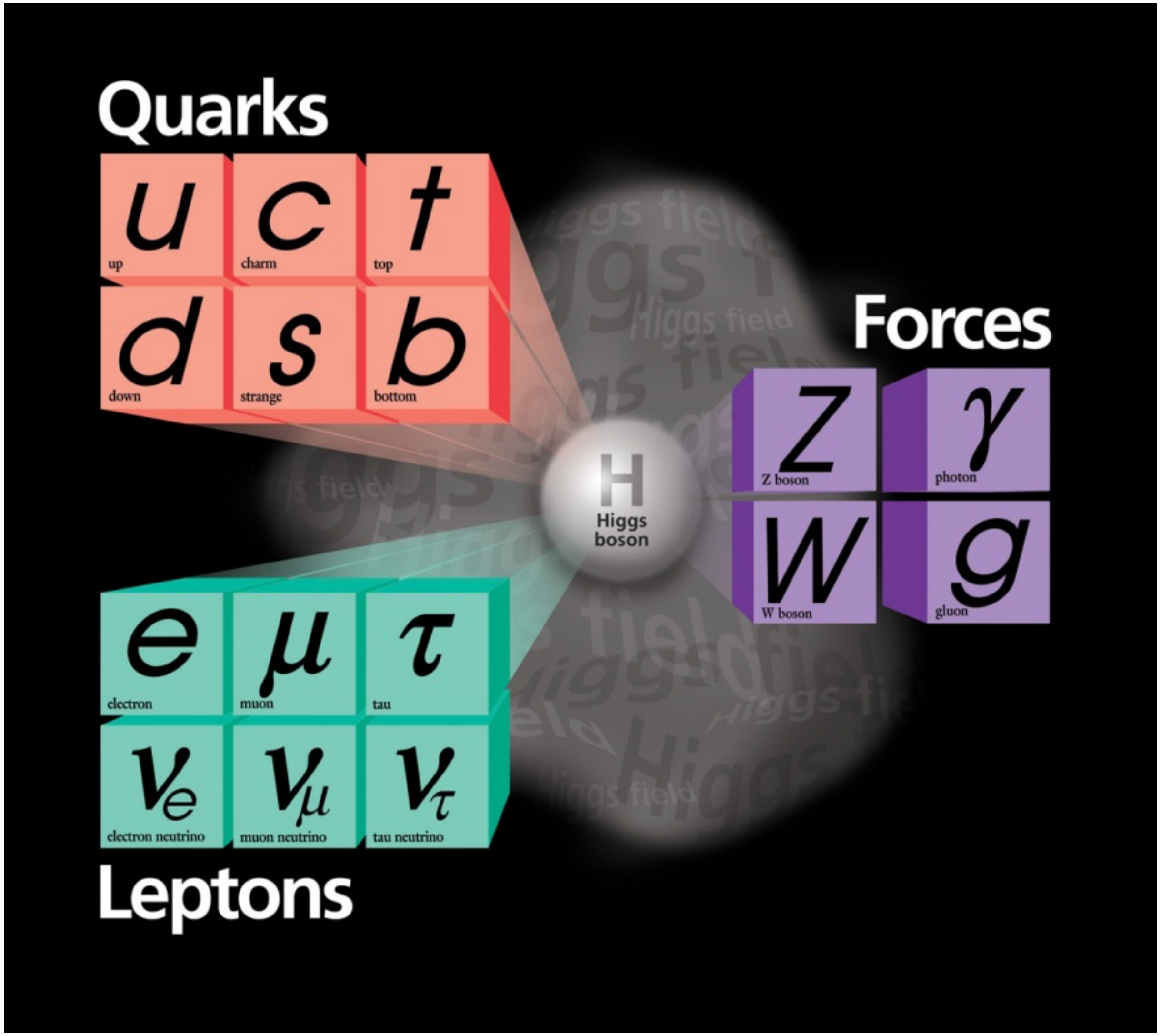
- The perturbation in energy levels depends on the symmetries of the material and the polarization of the electric field probing the material.

- The difference of the Hamiltonians perturbs in particular the energy expectation values of chiral materials.

- First order matrix elements in velocity gauge and length gauge differ at least at the several percent level for sub-femtosecond pulses.

- Differences between velocity gauge and length gauge should be particularly prominent for
  - → chiral materials
  - → strong fields
  - → short pulses

The matter which we know is described by the Standard Model of Particle Physics



Picture courtesy Fermilab

The matter which we know is described by the Standard Model of Particle Physics

$$
\begin{aligned}
\mathcal{L} = \sum_{(\nu,e)} \Bigg[& (\overline{\nu}_L, \overline{e}_L)\, \gamma^\mu \left(\mathrm{i}\partial_\mu + \frac{e}{\sin\theta} \boldsymbol{W}_\mu \cdot \frac{\boldsymbol{\sigma}}{2} - \frac{e}{2\cos\theta} B_\mu \right) \begin{pmatrix} \nu_L \\ e_L \end{pmatrix} + \overline{e}_R \gamma^\mu \left(\mathrm{i}\partial_\mu - \frac{e}{\cos\theta} B_\mu \right) e_R + \mathrm{i}\overline{\nu}_R \gamma^\mu \partial_\mu \nu_R \Bigg] \\
& + \sum_{(u,d)} \Bigg[(\overline{u}_L, \overline{d}_L)\, \gamma^\mu \left(\mathrm{i}\partial_\mu + \frac{e}{\sin\theta} \boldsymbol{W}_\mu \cdot \frac{\boldsymbol{\sigma}}{2} + \frac{e}{6\cos\theta} B_\mu + g G_\mu{}^a \frac{\lambda_a}{2} \right) \begin{pmatrix} u_L \\ d_L \end{pmatrix} \\
& \quad + \overline{d}_R \gamma^\mu \left(\mathrm{i}\partial_\mu - \frac{e}{3\cos\theta} B_\mu + g G_\mu{}^a \frac{\lambda_a}{2} \right) d_R + \overline{u}_R \gamma^\mu \left(\mathrm{i}\partial_\mu + \frac{2e}{3\cos\theta} B_\mu + g G_\mu{}^a \frac{\lambda_a}{2} \right) u_R \Bigg] \\
& - \frac{\sqrt{2}}{v_h} \sum_{(\nu,e)} \Bigg[(\overline{\nu}_L, \overline{e}_L) \begin{pmatrix} H^+ \\ H^0 \end{pmatrix} m_e e_R + \overline{e}_R m_e \left(H^{+*}, H^{0*} \right) \begin{pmatrix} \nu_L \\ e_L \end{pmatrix} \\
& \quad + (\overline{\nu}_L, \overline{e}_L) \begin{pmatrix} H^{0*} \\ -H^{+*} \end{pmatrix} \underline{M}_\nu \nu_R + \overline{\nu}_R \underline{M}_\nu^+ \left(H^0, -H^+ \right) \begin{pmatrix} \nu_L \\ e_L \end{pmatrix} \Bigg] \\
& - \frac{\sqrt{2}}{v_h} \sum_{(u,d)} \Bigg[(\overline{u}_L, \overline{d}_L) \begin{pmatrix} H^+ \\ H^0 \end{pmatrix} \underline{M}_d d_R + \overline{d}_R \underline{M}_d^+ \left(H^{+*}, H^{0*} \right) \begin{pmatrix} u_L \\ d_L \end{pmatrix} \\
& \quad + (\overline{u}_L, \overline{d}_L) \begin{pmatrix} H^{0*} \\ -H^{+*} \end{pmatrix} m_u u_R + \overline{u}_R m_u \left(H^0, -H^+ \right) \begin{pmatrix} u_L \\ d_L \end{pmatrix} \Bigg] \\
& - \left(\partial^\mu \left(H^{+*}, H^{0*} \right) + \mathrm{i} \frac{e}{\sin\theta} \left(H^{+*}, H^{0*} \right) \boldsymbol{W}^\mu \cdot \frac{\boldsymbol{\sigma}}{2} + \mathrm{i} \frac{e}{2\cos\theta} \left(H^{+*}, H^{0*} \right) B^\mu \right) \\
& \times \left(\partial_\mu - \mathrm{i} \frac{e}{\sin\theta} \boldsymbol{W}_\mu \cdot \frac{\boldsymbol{\sigma}}{2} - \mathrm{i} \frac{e}{2\cos\theta} B_\mu \right) \begin{pmatrix} H^+ \\ H^0 \end{pmatrix} - \frac{m_h^2}{2v_h^2} \left(H^+ H - \frac{v_h^2}{2} \right)^2 \\
& - \frac{1}{4} B_{\mu\nu} B^{\mu\nu} - \frac{1}{4} \boldsymbol{W}_{\mu\nu} \cdot \boldsymbol{W}^{\mu\nu} - \frac{1}{4} G_{\mu\nu}{}^a G^{\mu\nu}{}_a .
\end{aligned}
$$